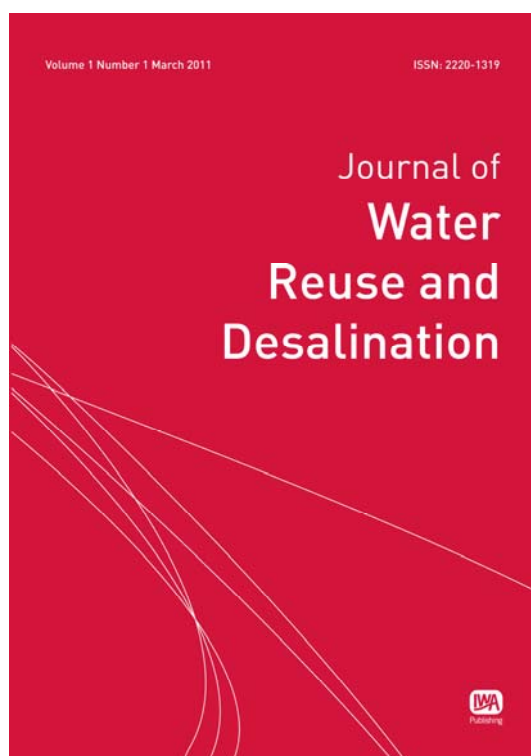


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Cleaning optimisation of reverse osmosis membranes used for wastewater reclamation

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ABSTRACT

Wastewater reclamation contributes to the preservation of conventional water resources and thus helps to ensure appropriate human development for future generations. Wastewater reclamation can be achieved through several technologies. One of the most common technologies is the tertiary treatment of urban municipal wastewater, which is often based on membrane technologies. Reverse osmosis is an effective separation technology for removing dissolved salts and low molecular weight organic compounds. However, membranes suffer from fouling, which directly reduces technical, environmental and economic feasibility of the process and hence of the reclamation plant. One of the strategies helpful to reduce fouling is the optimisation of the membranes' cleaning and maintenance. The aim of this work is to test the impact of the membrane cleaning protocol design on the recovery of the original properties of a reverse osmosis membrane used for several years in a wastewater reclamation plant in Spain. Furthermore, the work is focused on the validation of the adequacy of the most-common indicators used for assessing membranes' cleaning efficiency.

Key words | cleaning efficiency indicators, cleaning optimisation, fouling, membrane technology, reverse osmosis, wastewater reclamation

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LIST OF SYMBOLS

C_f	Feed conductivity ($\mu\text{S}/\text{cm}$)
C_P	Permeate conductivity ($\mu\text{S}/\text{cm}$)
J_{ac}	Permeate flux measured after cleaning ($\text{L}/\text{h m}^2$)
J_{bc}	Permeate flux measured before cleaning ($\text{L}/\text{h m}^2$)
R_{ac}	Salt rejection measured after cleaning
R_{bc}	Salt rejection measured before cleaning
RO	Reverse osmosis
SDS	Sodium dodecylsulphate
t	Cleaning duration (h)
T	Temperature ($^{\circ}\text{C}$)
TMP	Transmembrane pressure (bar)
v_f	Crossflow velocity (m/s)
ΔR	Rejection recovery (%)

INTRODUCTION

Around 300 km^3 of water are abstracted annually in Europe, which represents approximately 500 m^3 per capita/year

(European Environment Agency 2005). Around 32% of abstracted water is used for agriculture while industrial and domestic applications account for 53 and 15% of abstracted water, respectively (United Nations Development Program 2007). The population in the world is increasing at a rate of 80 million people/year. As a result, an increase of around 64 billion cubic metres of freshwater a year is required to satisfy human development needs (UNESCO 2009).

In Europe, the main source for freshwater production is surface water followed by groundwater (Dworak *et al.* 2007), which directly impacts on natural sources availability and water access guaranty. A survey completed in 2007 demonstrated that Spanish river basins led the classification of the most-stressed river basins among the 35 total surveyed European basins (European Environment Agency 2009). Alternative water resources such as rainwater harvesting, groundwater recharge, desalination and reclaimed water

reuse can be employed to reduce natural water sources use and therefore preserve their quality and quantity. Water reclamation can be achieved through the tertiary treatment of wastewater being the treatment configuration directly related to feed water quality, treated water requirements and site area availability among other factors. Membrane technologies are efficient and robust technologies useful to meet stringent reuse quality needs (Wintgens *et al.* 2005; Bixio *et al.* 2008; Baek & Chang 2009). Nevertheless, membranes are negatively affected by fouling phenomena created since the very beginning of their operation, which directly impacts on process efficiency, economy and environmental adequacy. Membrane fouling can be reduced and controlled through the optimisation of periodic cleaning actions performed during membranes' operation, apart from the optimisation of water pre-treatment and membrane operation, module configuration and hydrodynamics as well as on membrane material (Wolf *et al.* 2005; Tang *et al.* 2007; Strathmann *et al.* 2010). From an exploitation point of view, wastewater reclamation plant managers need to invest efforts in studying the optimal conditions for membrane pre-treatment, operation and cleaning in each particular application.

This work is focused on the study of the effect of the crossflow velocity (v_f), cleaning duration (t), cleaning strategy design (combination of recycling and soaking steps) on the recovery of the performance of a fouled reverse osmosis (RO) membrane element used for urban wastewater reclamation. Apart from testing the effect of cleaning variables on the recovery of the membrane properties, this work also deals with the study of the adequacy of the recovery of permeate flux and rejection as cleaning efficiency indicators.

MATERIALS AND METHODS

An 203 mm membrane element with a 4-year operation history was extracted from the lead position of the RO section of a wastewater reclamation plant in Spain to perform the cleaning study. The element corresponded to the LFC-1 model from Hydranautics and its autopsy revealed that fouling was mainly formed by biofouling (5.24×10^6 CFU/cm²). The standardised method ISO6222 was used to determine

aerobic bacteria content on the membrane fouling layer. For this, the deposit from a membrane sample was extracted with peptone solution and, after filtration and dilution, the obtained solution was incubated at 22 °C over 3 days using Water Plate Count Agar as cultivation media. Aerobic bacteria were subsequently counted. Permeability and salt rejection of the fouled element, measured through the filtration of 1,500 mg/L NaCl at 16 bar of transmembrane pressure (TMP), 25 °C of temperature (T) and 1 m/s of v_f , were 0.74 ± 0.08 L/h m²·bar and $92.2 \pm 1.2\%$, respectively. These values are 76 and 7% lower than virgin membrane specifications, respectively.

Sodium hydroxide (NaOH) containing 0.03% sodium dodecylsulphate (SDS) at pH 12 was used as cleaning solution for all the experiments because basic formulations are known to be efficient for the removal of organic and biological fouling from membranes (Al-Amoudi & Lovitt 2007). Moreover, in a preceding cleaning study performed with the same membrane element as in the present work it was concluded that, in terms of chemical composition, the most effective cleaning formulation had the above mentioned composition (Bernat *et al.* 2011).

A laboratory-scale membrane crossflow filtration rig for testing flat-sheet polymeric membranes was used for performing the cleaning experiments. The unit, schematised in Figure 1, was composed of a temperature-controlled feed tank (1), a pump equipped with an adjustable speed drive (2), a flat-sheet filtration cell (4) manufactured by GE Osmonics (model SEPA CF II), a backpressure valve (5) and two purge valves (3). Two pressure gauges were installed (P1 and P2) to monitor TMP. A BEL Engineering balance was installed in the permeate outlet to monitor permeate mass and thus obtain permeate flux. A lab-scale

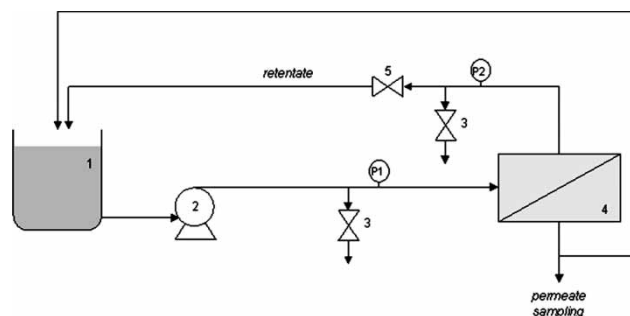


Figure 1 | Membrane filtration unit.

multimeter (Crison) was used to analyse permeate and feed pH and conductivity.

Flat-sheet membrane samples were withdrawn from the full-scale element selected to perform cleaning experiments. A new membrane coupon was used for each cleaning condition. Before starting a cleaning test, the fouled membrane was characterised in terms of permeate flux and salt rejection. With this aim, a 1,500 mg/L NaCl solution was filtered at 16 bar of TMP, 25 °C and 1 m/s of v_f . Permeate flow as well as feed and permeate conductivity (C_f and C_p , respectively) were continuously monitored so that, after reaching the steady state, fouled membrane permeate flux (J_{bc}) and rejection (R_{bc}) could be calculated. J_{bc} was calculated from permeate flow, membrane surface area and TMP whilst R_{bc} was calculated according to Equation (1).

$$R_{bc}(\%) = 100 \times \left(1 - \frac{C_p}{C_f}\right) \quad (1)$$

After the fouled membrane characterisation, the cleaning experiment started. For this, 2 L of cleaning solution were placed in the feed tank and after the system had reached the temperature set point (25 °C), the cleaning started. All the cleaning tests were performed at 2 bar of TMP and 25 °C and they were performed in total recycling mode (unless

otherwise specified) so both permeate and retentate were recycled back to the cleaning solution vessel. Once the cleaning experiment was completed, cleaned membrane characterisation was carried out following the same protocol as for fouled membrane characterisation. Cleaned membrane characterisation was performed to calculate cleaned membrane flux (J_{ac}) and salt rejection (R_{ac}). Cleaning efficiency was determined through the permeate flux gain (J_{ac}/J_{bc}) and by comparing R_{ac} to R_{bc} .

RESULTS AND DISCUSSION

Effect of crossflow velocity on basic cleaning efficiency

In this section, the effect of the crossflow velocity applied during the cleaning sequence on cleaning efficiency is discussed. The experiments were performed for 2 h working at recycling mode. As it can be observed in Figure 2, J_{ac}/J_{bc} remains practically constant regardless of v_f applied. Similar results were obtained by Bartlett *et al.* (1995) in a cleaning study of microfiltration membranes with NaOH. A raise of the crossflow velocity of the cleaning solution is expected to increase the shear rate close to the membrane surface and therefore improve foulants' removal. However, an increase of this variable reduces the contact time

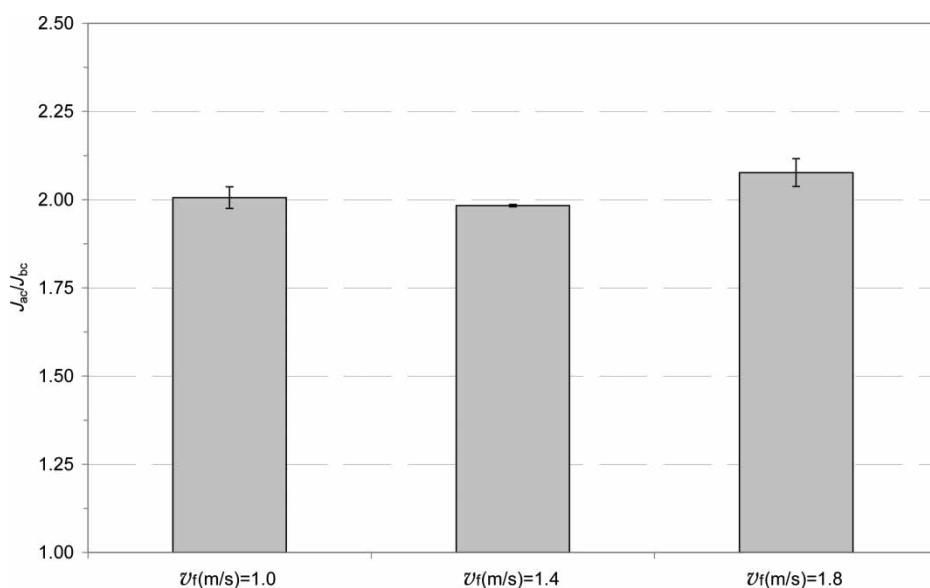


Figure 2 | Influence of crossflow velocity on permeate flux gain. NaOH+0.03% SDS; pH = 12; t = 2 h (recycling); TMP = 2 bar; T = 25 °C.

between the compounds forming the fouling deposit and cleaning agent, which can worsen the fouling removal efficiency achieved. The balance between shear rate and contact time thus determines the impact of v_f on cleaning efficiency and, in the case presented, no macroscopic effect is observed on permeate flux gain when v_f is modified.

The effect of v_f on the membrane rejection recovery is shown in Figure 3. Membrane salt rejection was partially recovered regardless of v_f . A minimum value in rejection gain was observed at the intermediate v_f tested. This could indicate that at these conditions the contact time between chemicals and foulants was too short and the shear effects on membrane surface neighbourhoods are minimal. Figure 3 also shows that when v_f increases, shear effects improve rejection but not to the same extent as the lowest v_f values.

In a previous study carried out by Bernat *et al.* (2011) with the same membrane element as that in the present work, it was evidenced that membrane rejection recovery was not an appropriate cleaning efficiency indicator because it was strongly influenced by the fouled membrane rejection. Membrane rejection behaviour can thus screen the effect of v_f , and of other cleaning variables on rejection recovery, as demonstrated in the above mentioned publication. The factors conducting to the statement of the previous hypotheses can be thus altered by the rejection effect and

be inexact or even uncertain. The inadequacy of membrane rejection recovery as a cleaning efficiency indicator for the selected membrane is demonstrated in the last section.

Effect of cleaning duration and cycles' design on cleaning efficiency

The influence of the cleaning duration and of the distribution of the cleaning time in recycling and soaking sequences is discussed in this section. With the aim to test the aforesaid variables' impact on cleaning efficiency, experiments designed with only recycling or soaking as well as with the combination of recycling and soaking stages were carried out. Recycling refers to the continuous crossflow recirculation along the membrane lumen of the chemical formulation tested. Instead, soaking is carried out without any movement of the cleaning solution. When the efficiency of combined recycling and cleaning studies was evaluated, recycling was always the first step followed by the static soaking stage.

The cleaning formulation used for all the experiments presented in this section was based on NaOH+0.03% SDS at pH 12. TMP, v_f and T were kept constant at 2 bar, 1 m/s and 25 °C during the cleaning experiments, respectively. No pressure and v_f were obviously applied in soaking-

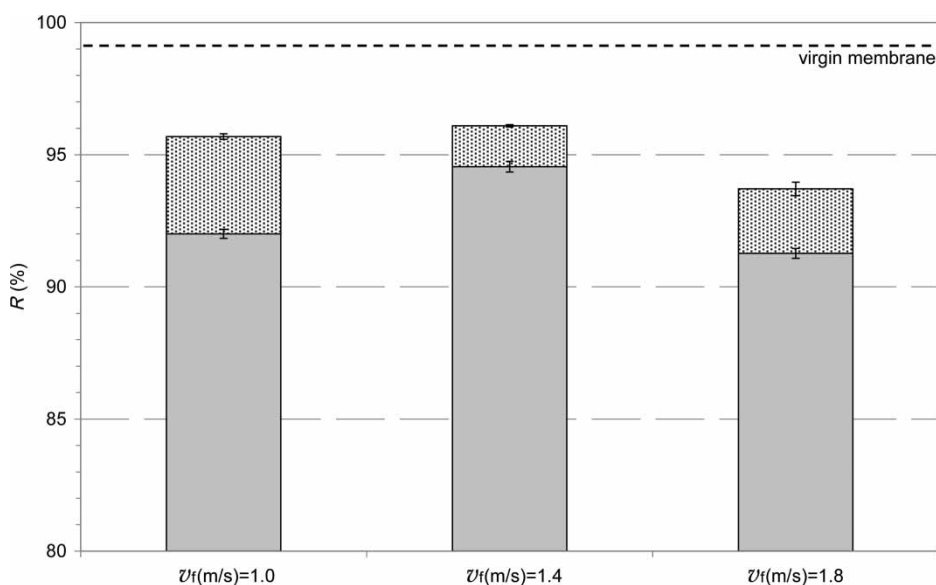


Figure 3 | Influence of crossflow velocity on membrane rejection. NaOH + 0.03% SDS; pH = 12; t = 2 h (recycling); TMP = 2 bar; T = 25 °C. Solid bars represent fouled membrane rejection and textured bars cleaned membrane rejection.

including experiments. Figure 4 shows the effect of the cleaning duration and cleaning cycles' design on permeate flux decline. Grey bars represent the experiments performed at recycling mode and thus without the combination of soaking sequences. The tests were performed at a total duration ranging from 0.5 to 6 h and Figure 4 demonstrates that between 0.5 and 2 h, the higher the cleaning duration the greater the cleaning efficiency achieved. However, beyond 2 h, a cleaning duration increase does not result in an improvement of the permeate flux gain. Between 0.5 and 2 h, an increase on the cleaning duration, which intrinsically results into a higher contact time between the cleaning agent and the fouling layer, increases the amount of deposit layer removed from the membrane and thus improves cleaning efficiency. Beyond 2 h, regardless of the cleaning duration, J_{ac}/J_{bc} remains constant at around 2. This behaviour may be explained by the fact that cleaning activity is probably consumed during the first 2 h of cleaning, by the presence of non-removable fouling and/or due to the ageing of the membrane studied.

Cleaning tests consisting of only soaking, which are represented in black bars in Figure 4, give lower J_{ac}/J_{bc} than when recycling was performed at the same conditions. This can be attributed to the fact that, when soaking is performed, lower contact and diffusion of the cleaning agent through the deposit layer occurs. Figure 4 shows that an

increase in the soaking duration improves the permeate flux gain. The higher recovery of the permeate flux achieved at high soaking duration can be explained by the above mentioned diffusion/contact time mechanisms. As cleaning agent interaction with fouling components is lower for soaking than recycling, the consumption of the activity of the agent occurs faster for the former than for the latter.

Combined cleaning cycles (including recycling and soaking) do not exhibit any improvement of the cleaning efficiency, as can be observed in Figure 4. The presence of soaking and recycling steps in the same cleaning experiment improves the cleaning efficiency when compared to the soaking-alone experiments at the same total duration. However, when J_{ac}/J_{bc} of combined cycles is compared to J_{ac}/J_{bc} of the experiments consisting of only a recycling sequence at the same total duration, a decrease in the cleaning efficiency is achieved. This also demonstrates that the removal of compounds present in the fouling layer is greater when recycling is performed. As explained above, at recycling conditions, a higher contact of the cleaning agents with and penetration through the fouling deposit occurs owing to the favourable conditions created by the crossflow mode at the membrane skin layer.

The effect of the cleaning sequence design on membrane rejection recovery can be observed in Figure 5. The results represented in grey and white bars show that in all the

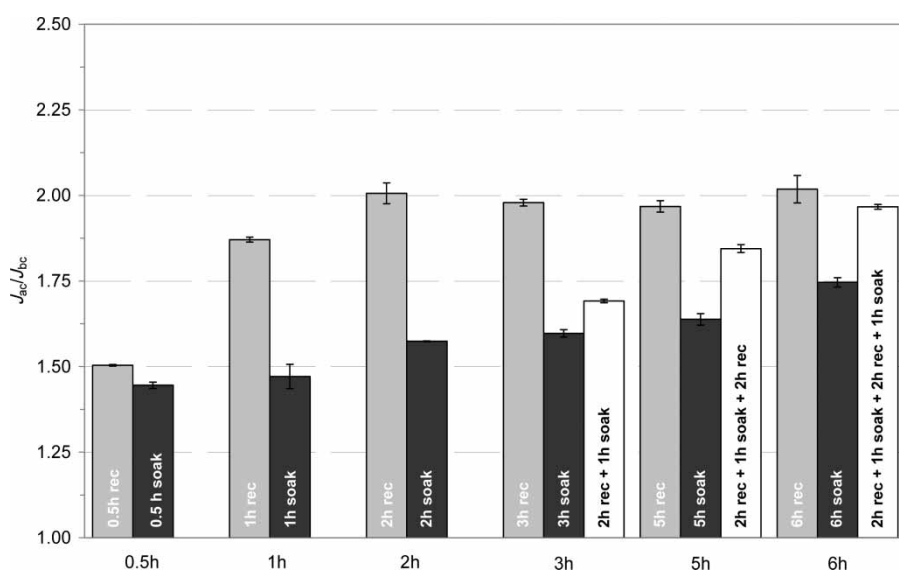


Figure 4 | Influence of cleaning duration and cycles' design on permeate flux gain. NaOH + 0.03% SDS; pH = 12; $v_f = 1$ m/s; TMP = 2 bar; $T = 25$ °C. Grey bars represent recycling experiments, black bars soaking experiments and white bars combined recycling and soaking experiments.

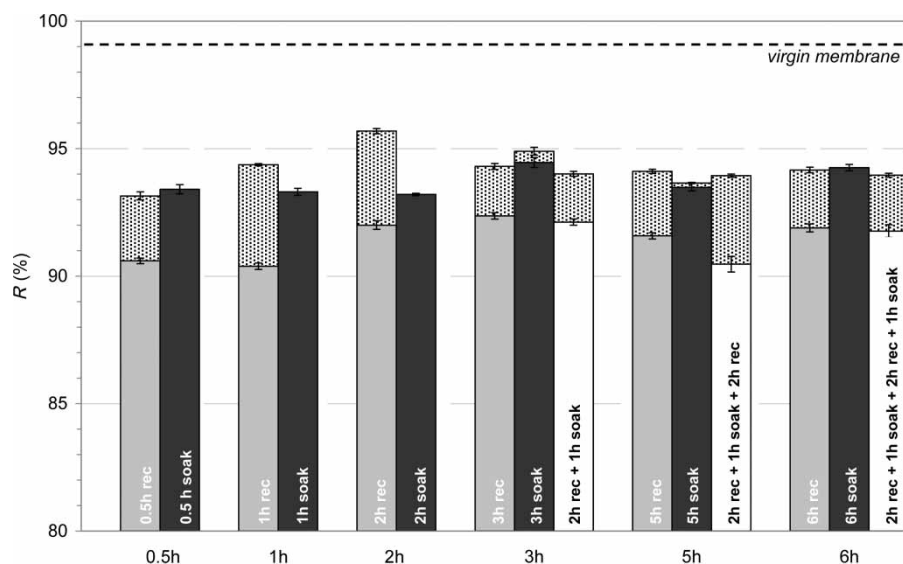


Figure 5 | Influence of cleaning duration and cycles' design on membrane rejection. NaOH+0.03% SDS; pH = 12; $v_f = 1$ m/s; TMP = 2 bar; $T = 25$ °C. Grey bars represent recycling experiments, black bars soaking experiments and white bars combined recycling and soaking experiments.

recycling-containing cleaning experiments an improvement on the permeate flux was achieved compared to soaking-alone conditions. The same figure shows that soaking alone is not capable of restoring membrane rejection. The previous observations can be explained by the same mechanism as for the permeate flux gain response. As soaking mode restrains cleaning agents' interaction with fouling compounds, its cleaning efficiency is markedly lower than for recycling. It is worth mentioning that, as observed in the experiments presented in the previous section, the unclear response of rejection after cleaning with cleaning conditions variations can be explained by the inadequacy of this parameter as a cleaning efficiency indicator. The following section deals with the assessment of the adequacy of permeate flux gain and rejection as cleaning efficiency indicators for the membrane studied.

Cleaning efficiency indicators' adequacy

The assessment of the optimal cleaning conditions for removing fouling from used RO membranes passes through not only the determination of the cleaning parameters influence on cleaning efficiency but also the evaluation of the adequacy of the cleaning efficiency indicators employed. In this work, as exposed in the previous sections, permeate flux gain and rejection recovery have been selected as

cleaning efficiency indicators. The results discussed in the preceding sections demonstrate that the permeate flux gain reflects the effects of changing the cleaning experimental conditions contrarily to what happens with the membrane salt rejection recovery. In order to demonstrate the adequacy of both indicators for cleaning efficiency determination, J_{ac}/J_{bc} and rejection recovery (ΔR) values have been plotted against the fouled membrane properties (J_{bc} and R_{bc}) in Figures 5 and 6, respectively. The values plotted in the above mentioned figures are those obtained in the experiments discussed in the preceding sections as well as those published by Bernat *et al.* (2011) for the same membrane element. The experimental conditions thus

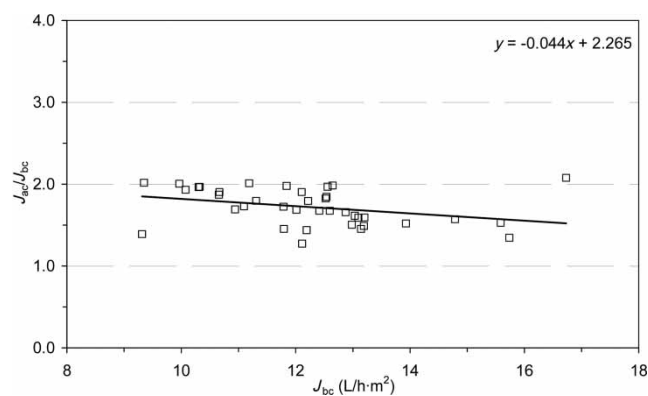


Figure 6 | Permeate flux gain dependence on fouled membrane permeate flux.

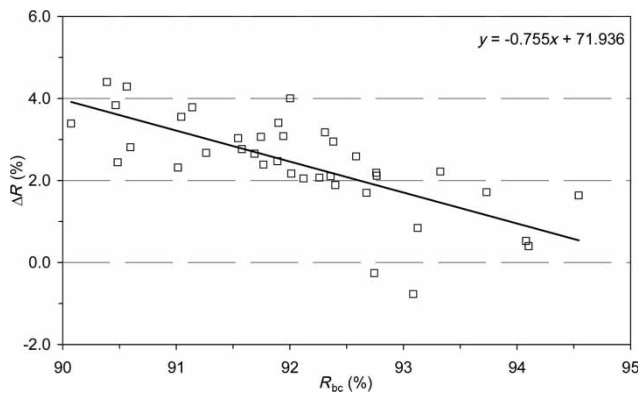


Figure 7 | Rejection recovery dependence on fouled membrane rejection.

differ among experiments so that the influence of the initial status of the fouled membrane, which has been characterised through J_{bc} and R_{bc} , on the membrane cleaning efficiency measured can be assessed regardless of the cleaning conditions.

Figure 6 demonstrates that the permeate flux gain is practically independent of the fouled membrane permeate flux because the regression slope calculated from the J_{ac}/J_{bc} versus J_{bc} plot is close to zero (-0.044). Thus, it can be concluded that J_{ac}/J_{bc} is an appropriate cleaning efficiency indicator for the membrane studied as it only depends on the cleaning conditions applied and not on the fouled membrane conditions.

In contrast to the J_{ac}/J_{bc} behaviour, rejection recovery is significantly affected by the fouled membrane salt rejection. Figure 7 evidences that ΔR is dependent on R_{bc} because the slope of the regression of ΔR versus R_{bc} is -0.755 . This clearly indicates that rejection is not an appropriate and reliable cleaning efficiency indicator for the membrane studied because it depends not only on the cleaning conditions but also on the fouled membrane rejection. Therefore, rejection can only be used to determine fouled and cleaned salt selectivity of the membrane but not to assess the efficiency of cleaning protocols.

CONCLUSIONS

Cleaning efficiency of an RO membrane used for wastewater reclamation depends on cleaning duration and cleaning cycles' design. An increase in the total cleaning

duration improves cleaning efficiency up to a certain value beyond which further improvements are not observed because of the probable total consumption of the cleaning agent, riddance of all removable fouling and effects of the membrane ageing. Recycling sequences more efficiently restore membrane performance than soaking steps because of the favourable hydrodynamic conditions occurring in recycling steps, which allow cleaning agents to interact to a higher extent with the compounds forming the fouling layer and easily diffuse through it. At fixed total cleaning duration, the combination of recycling and soaking steps results in an improvement of the cleaning efficiency achieved compared to soaking-alone tests and into an efficiency diminishment compared to recycling-alone experiments. This can be also attributed to the advantageous effect of the higher contact of the cleaning agents with the fouling layer occurring at recycling conditions.

Tangential velocity during cleaning does not exhibit any effect on the membrane restoring achieved. This behaviour can be explained by the balance existing between the shear rate at the active layer neighbourhoods and the contact time between the cleaning agent and the fouling layer substances. A crossflow velocity raise results in both an improvement of the hydrodynamic conditions at the membrane surface surroundings and a worsening of the contact degree between fouling components and cleaning agent. Therefore, although no changes on the cleaning efficiency are macroscopically observed when crossflow velocity is modified, phenomena resulting from the velocity adjustment can indeed occur.

An appropriate indicator for assessing cleaning efficiency for the studied membrane has been demonstrated to be the permeate flux gain achieved during cleaning. Membrane rejection recovery has been found to be an inappropriate and therefore unreliable cleaning efficiency indicator for the RO membrane selected due to its direct link to the original salt rejection of the fouled membrane element. This conclusion undoubtedly evidences the importance of focusing on membrane cleaning studies, apart from testing the effect of cleaning variables on cleaning efficiency, on the selection of the most accurate and reliable cleaning efficiency indicators. This will avoid extracting inaccurate conclusions from

laboratory-scale cleaning studies that can be afterwards used to adapt cleaning protocols at full-scale plants.

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